Experimental Tests of the New Paradigm for Laser Filamentation in Gases

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Since their discovery in the mid-1990s, ultrafast laser filaments in gases have been described as products of a dynamic balance between Kerr self-focusing and defocusing by free electric charges that are generated via multi-photon ionization on the beam axis. This established paradigm has been recently challenged by a suggestion that the Kerr effect saturates and even changes sign at high intensity of light, and that this sign reversal, not free-charge defocusing, is the dominant mechanism responsible for the extended propagation of laser filaments. We report qualitative tests of the new theory based on electrical and optical measurements of plasma density in femtosecond laser filaments in air and argon. Our results consistently support the established paradigm.

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Since the first experimental observations of filament propagation in air in the mid-1990s [1] the field of ultrashort laser filamentation in gases has matured and enabled various technologically important applications including few-cycle optical pulse generation [2], THz generation [3], remote sensing [4], laser-triggered lightning [5], and even laser-induced cloud formation [6]. In parallel with experimental progress, the theory and simulation of laser filamentation has advanced greatly, allowing for example, for modeling of the supercontinuum generation and third-harmonic generation that accompany filament propagation [7]. Given this tremendous progress, it is still fair to say that the basic paradigm of filament formation has remained unchanged from the mid-1990s. Namely, the self-focusing collapse of a high peak power input pulse in a gas due to the nonlinear Kerr effect is arrested by the defocusing action of free electrons that are generated on the beam axis via multi-photon ionization [8],[9].

The established "old" paradigm has been recently challenged by a suggestion that Kerr self-focusing in various gases can saturate and even become defocusing as the optical intensity is increased [10]. This suggestion represents a radical departure from our past understanding of filament formation. Although the saturation of the Kerr effect has been suggested to be a factor in laser filamentation [11–13], it was never assumed to be the dominant one. If proven valid, the "new" paradigm would have profound implications in filamentaion science. On one hand, the sign reversal of the Kerr effect would enable plasma-free filament propagation, which could be much longer ranged, owing to the much reduced energy losses into ionization compared to that in the established filamentaion scenario [14]. In addition, various nonlinear optic conversion processes inside filaments, such as third and fifth harmonic generation, could become very efficient [15]. On the other hand, applications that rely on the presence of plasma inside filaments would suffer.

The goal of this paper is to present comprehensive experimental tests of the new paradigm. Our results show that defocusing by free electric charges is certainly strong enough to balance Kerr self-focusing in femtosecond laser filaments, thus supporting the established paradigm. Since our experiments are designed such that they measure qualitative, not quantitative differences between the predictions of the established and the new theories, our conclusions should not be affected by the variability of the material parameters. We stress that our findings do not imply that the higher-order nonlinear terms advocated in [10] are nonexistent. Our conclusion is that, even if the higher-order nonlinear terms do exist, the free-charge generation and the associated defocusing in a filament set in early enough to mask their effect, thus rendering them inoperative.

Our first test is based on the electrical conductivity of filaments. Direct and quantitative measurements of the density of charges generated through laser filamentation are problematic owing to both the very high intensity inside filaments and short lifetime of the generated plasma. Accordingly, the reported values of plasma density inside filaments vary by several orders of magnitude (see, for example, [8]). Furthermore, plasma density inside filaments has been shown to be strongly dependent on the external focusing conditions [16]. Any attempt to validate or disprove the new filamentation paradigm based on a quantitative measurement of electric charges generated through filamentation would be very challenging. Luckily, for a particular pair of gases, namely air and argon, the old and new filamentation theories predict vastly different values of the generated free-charge density thus offering an opportunity to validate the new theory based on a semi-qualitative measurement.

Indeed, by examining the experimental data on the nonlinear refractive index as a function of the intensity of light that have been reported for various gases in [10], one notices that the curves for air and argon are essentially identical. This close similarity extends well into the intensity range in which, according to the new filamentation theory based on this very data, the Kerr effect changes sign and becomes defocusing. At the same time, the ionization potential of argon is considerably higher than that of air. For laser pulses at 800 nm that we use in our experiments, it takes 11 photons to ionize argon, while ionizing oxygen, one of the major constituents of air, requires only 8 photons.

According to the above considerations, if the filament is indeed stabilized by sign reversal of the Kerr effect (new theory), then filaments generated in air and argon under otherwise identical conditions would have very similar spatial intensity distributions. Plasma, which is a bystander in this scenario, would be generated in proportion to the multi-photon ionization rate. In that case, one would expect a lot more plasma to be generated through filamentation in air than in argon.

On the other hand, if plasma defocusing is the major stabilizing mechanism in filaments and the Kerr effect is always positive (old theory), then filamentation in air would generate less plasma than that in argon. In this scenario, the higher ionization potential of argon would cause plasma generation to be initiated later in the focusing cycle. The filament in argon would be thinner than in air. It would take more plasma to defocus this thinner filament because it has higher peak intensity and associated stronger self-focusing.

Numerical simulations of filamentaion with 1 mJ, 30 femtosecond-long pulses in air and argon reported in [14] agree with the above qualitative assessment. For the case of the new theory, these simulations predict about ten times as much plasma in air as in argon. For the case of the old theory, the simulations predict about twice as much plasma in argon as in air.

To experimentally verify the validity of the above predictions, we initiate filaments in air and argon under atmospheric pressure, using 35 femtosecond-long laser pulses with 800 nm center wavelength and various pulse energies. The laser beam with a 1 cm diameter is weakly focused with a lens of about 190 cm focal length, making the focusing conditions similar to those used in the simulations reported in [14]. To measure linear plasma density, in arbitrary units, we use a standard capacitive plasma probe schematically shown in the top portion of Figure 1 and described in detail elsewhere [17]. In this particular case, the probe has 1 cm × 1 cm square electrodes separated by 1.5 mm. The DC voltage applied to the electrodes is 200 V. The results of the plasma density measurements in air and in argon are shown in the bottom part of Figure 1. It is evident that in all cases,

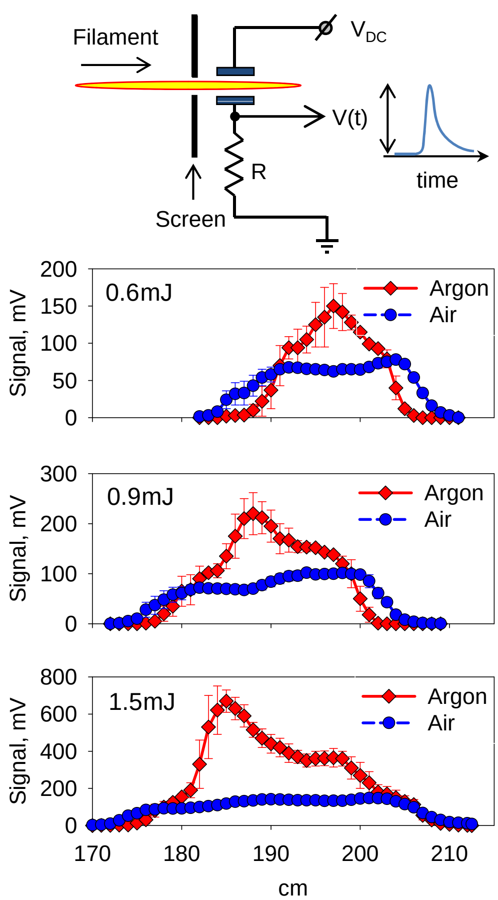

FIG. 1: Top panel: Schematic of the capacitive plasma probe. Bottom panels: Experimental data for plasma density generated through filamentation in air and argon under identical conditions, for three different values of pulse energy.

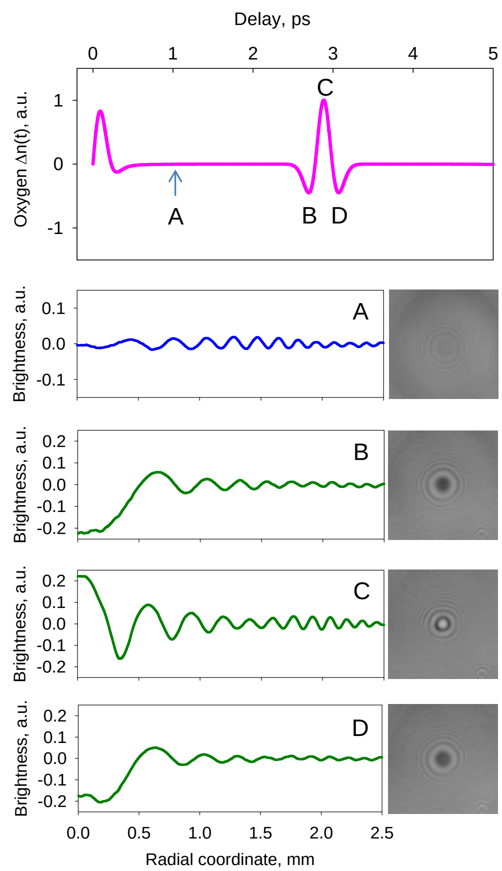

FIG. 2: Top panel: Calculated rotational Raman response for molecular oxygen. Bottom panels: Measured diffraction patterns and corresponding radial intensity profiles of the probe pulse in air for four particular values of the pump-probe delay as indicated by letters A – D on the revival curve. Point A does not overlap with any revival feature of either oxygen or nitrogen.

plasma density generated in argon is higher than in air, in agreement with the established filamentation theory.

Owing to the semi-qualitative nature of the above test, the end result confirming the validity of the established theory appears sufficiently conclusive. However, the weakness of this test is related to the fact that the plasma probe that we use is gas specific. Only a small fraction of the generated charges is captured by the probe for each individual laser pulse. The measured electrical signal is related not only to the probe geometry and plasma density in the filament, but also, in some nontrivial way, to the free-electron mobility and recombination rate in a particular gas. Although these parameters are similar for air and argon, they are certainly not identical for the two gases [18]. Thus strictly speaking, using this plasma probe for quantitative comparison of filament properties in different gases is not entirely justified.

Our alternative test of the new theory is based on the diffraction of a collimated probe beam on a plasma channel generated through filamentation. The experimental setup is similar to the one reported in [19]. Filaments in air and in argon are generated under the same conditions as in the conductivity experiments described above, but the pulse energy is fixed at 0.9 mJ. (At higher pulse energies in argon we observe the onset of bright conical emission that interfered with the measurements.)

A fraction of the incident 800 nm beam is split-off and frequency doubled in a 200 μm-thick, 1 cm $\times$ 1 cm BBO crystal and used as a collimated probe beam. We estimate that the probe pulse has a duration of less than 50 femtoseconds. The energy of the probe pulse is about 10 μJ, thus the probe propagates in a linear regime. The polarization of the probe is controlled by a half-wave plate, and in the experiments reported here polarizations of the pump and probe pulses are parallel.

The collimated probe beam, after recombination with the pump in a dichroic mirror, propagates collinearly with the pump. The time delay between the pump and the probe pulses is controlled via a mechanical delay line. The probe beam diffracts on the pump-generated filament and the resulting diffraction pattern is photographed by a CCD camera placed at a distance of 75 cm from the center of the filament. The 800 nm pump light incident on the camera is blocked by a color-glass filter. In this setup, the probe pulse non-destructively samples index changes that are experienced by the intense pump pulse as it undergoes filamentation.

In air, the nonlinear response of the medium to an ultrashort optical pulse is composed of three components: The instantaneous Kerr effect, a delayed rotational Raman response which comes in the form of periodical revivals, and a free-charge defocusing which lasts for several hundred picoseconds. For the purposes of differentiation between the old and new filamentation theories, our goal here is to establish a relationship between the strengths of Kerr self-focusing and free-charge de-focusing.

The Raman component of the medium response can be calculated [20]. The result of this calculation for the refractive index change due to the oxygen content of air is shown, in arbitrary units, in the top part of Figure 2. The bottom part of the Figure shows examples of experimentally recorded diffraction patterns and corresponding radial intensity distributions of the 400 nm probe beam, for four particular values of the pump-probe delay. The distributions shown are obtained by digitizing the corresponding diffraction patterns along the line passing through the center of the pattern, followed by subtraction of the profile obtained without filament (with the 800 nm beam blocked).

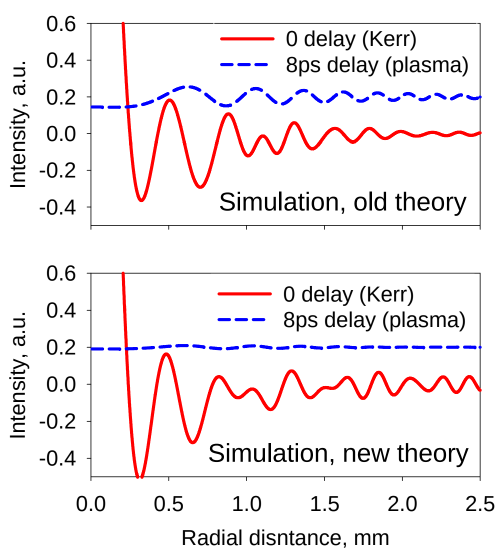

FIG. 3: Radial intensity distributions of the probe pulse in argon that have been numerically simulated using old theory (top), and new theory (bottom), for two different values of the pump-probe delay. Profiles for 8 picosecond delay are offset upwards, for clarity.

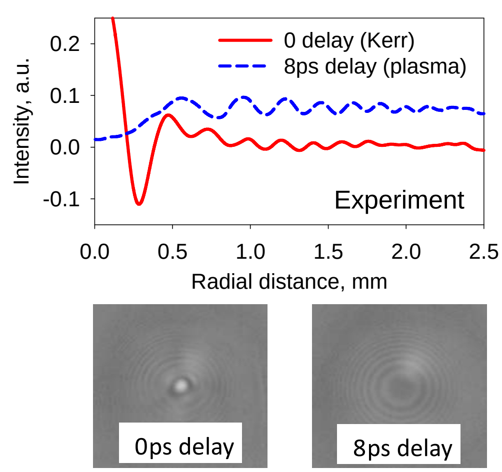

FIG. 4: Experimental data for filamentation in argon under same conditions as those used in simulations shown in Figure 3. Profile for 8 picosecond delay is offset upwards. Agreement with the old theory is evident.

In all cases, the negative (defocusing) swings of the revival curve produce patterns with dark centers, while features corresponding to the positive (focusing) swings produce patterns with bright centers. Patterns corresponding to the rotational revivals due to molecular nitrogen show the same tendency. From this data we deduce an estimate of the temporal resolution attainable in our experiments. Delays between successive focusing-defocusing features on the revival curve (e.g. between points B and C in Figure 2) are about 200 femtoseconds. Since our technique clearly distinguishes between these features, the temporal resolution of our experiments is substantially better than 200 femtoseconds. The ultimate temporal resolution that can be achieved is limited by the duration of the probe pulse and by the walk-off between the pump and probe pulses due to the group-velocity dispersion over the length of the filament, which is about 20 femtoseconds in this case.

Pattern A in Figure 2 is obtained for a 1 picosecond delay between pump and probe pulses. Neither Kerr effect nor any revival feature for either oxygen or nitrogen affects the propagation of the probe pulse in that case, thus this pattern is produced by plasma defocusing. As expected, the pattern has a dark center. However, quantifying the strength of plasma defocusing and relating it to that of self-focusing based on the data obtained for air is problematic because the Kerr response at the zero time delay is masked by the Raman response.

To establish a quantitative relationship between focusing action of the Kerr effect and plasma defocusing, we conducted the same diffraction experiment as above in argon atmosphere. Argon is an atomic gas, therefore its nonlinear optical response is free from the rotational Raman component inherent to molecular gases such as air.

Note that changes in refractive index due to the Kerr effect are proportional to the instantaneous optical intensity. Therefore the transverse geometry of the scatterer that the probe beam diffracts from, when pump and probe pulses overlap in time, follows the spatial intensity profile of the pump. On the other hand, the spatial profile of the generated plasma is proportional to a high power of intensity. Accordingly, the size of the defocusing scatterer due to the plasma is smaller than that of the intensity distribution of the pump. As a result, diffraction patterns generated by plasma should be, in general, wider compared to those generated by the pump-induced Kerr focusing.

Through extensive numerical simulations of our experiment we found that quantitative comparison between self-focusing due to the Kerr effect and the defocusing action of plasma has to be drawn based on the shape of the peripheral parts of the diffraction patterns, not their centers. The results of numerical simulations for the intensity distribution of the probe beam based on the old and new filamentaion theories are shown in Figure 3. In the case of the old model, the oscillations in the peripheral parts of the patterns corresponding to the zero and large pump-probe delays have comparable contrasts. On the contrary, the peripheral oscillation due to plasma defocusing in the case of the new model is essentially invisible. This again represents a qualitative difference between the predictions of the two theories.

Experimental results for the case of filamentation in argon under the conditions used in the above simulations are shown in Figure 4. As in the case of filamentation in air, the intensity profiles are obtained by digitizing the photographed diffraction patterns along the line passing through the pattern's center and subtracting the profile obtained with the pump blocked. Agreement with the simulation based on the established theory is evident.

In conclusion, we have conducted two independent experiments testing the new filamentation paradigm, according to which a sign reversal of the Kerr effect is the dominant physical process that stabilizes laser filamentation in gases. Both tests consistently disqualified the new paradigm and supported the established theory that treats free-charge defocusing as the dominant stabilization mechanism.

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